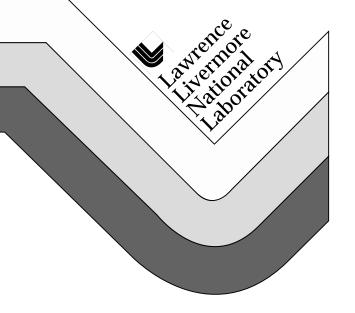# TOWARD IMPROVED PHOTON-ATOM SCATTERING PREDICTIONS

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TOWARD IMPROVED PHOTON-ATOM SCATTERING PREDICTIONS[*]

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Photon-atom scattering is important in a variety of applications, but scattering from a composite system depends on the accurate characterization of the scattering from an isolated atom or ion. We have been examining the validity of simpler approximations of elastic scattering in the light of second-order S-matrix theory. Partitioning the many-body amplitude into Rayleigh and Delbrück components, processes beyond photoionization contribute. Subtracted cross sections for bound-bound atomic transitions, bound pair annihilation, and bound pair production are required in anomalous scattering factors for: 1) convergence of the dispersion integral; 2) agreement with predictions of the more sophisticated S-matrix approach; 2) satisfying the Thomas-Reiche-Kuhn sum rule. New accurate tabulations of anomalous scattering factors have been prepared for all Z, for energies 0-10,000 keV, within the independent particle approximation (IPA) using a Dirac-Slater model of the atom. Separately, experimental atomic photoabsorption threshold information has been used to modify these IPA predictions for improved comparison with experiment.

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1. Introduction

Scattering of photons by atoms, molecules and solids is an important method for obtaining information about structural properties of materials. Scattering is also important in simulations of radiation transport in complex systems, for example, in an ICF plasma, an x-ray optical element like an x-ray mirror, or a medical diagnostic device. But, scattering from a composite system depends on the accurate characterization of the scattering from the basic unit, an isolated atom or ion. In this report, we note the progress that has been made toward accurate, reasonably simple elastic photon-atom scattering predictions based on use of modified relativistic form factors and angle-independent anomalous scattering factors.

The S-matrix approach [1] is at present the best first-principles calculation of Rayleigh scattering within independent particle approximation (IPA – in which the scattering from each electron is computed separately). Agreement with S-matrix predictions in comparisons with experimental results is found to exist over a wide range of photon energies [2], not too near photoeffect thresholds. The method requires a considerable amount of computation time which makes extensive systematic computation more difficult to perform. Recently, the validity of simpler approximations in the prediction of elastic [3] scattering has been examined in the light of numerical calculations via second-order S-matrix theory.

The form-factor approximation (FF) or its improved version, the modified relativistic form factor (MF), is a widely used simpler but useful approach. Form-factor approximations are small-momentum-transfer, high-energy approximations and one

expects substantial deviations for photon energies near and below the photoeffect thresholds, as well as for heavy elements and large momentum transfers. The MF, which contains important electron-binding corrections, produces better high-energy results for heavy elements, and more importantly, gives the correct relativistic high-energy limit for forward-angle scattering [4]. Extensive tabulations of FF [5, 6] and MF [7] are readily available for all elements.

Anomalous scattering factors (ASF) are a useful approach for forward-angle scattering, obtained using dispersion relations (relating the real and imaginary parts of the forward scattering amplitude) and the optical theorem (relating the imaginary forward-angle scattering amplitude to the total photo-atom cross section). Utilizing only bound-free photoabsorption data, several tabulations of anomalous scattering factors are available [8-10]. More recently, some discussion of problems with existing ASF tabulations has appeared [4, 11, 12].

Anomalous scattering factors can be combined with FF or MF by taking ASF as (an assumed angle-independent) deviation from form-factor approximation at all angles. This approach does not generally give the same accuracy as the S-matrix calculation, and the angle-independent approximation is not adequate at large momentum transfers, but it improves FF or MF substantially to give very good results at low photon energies.

2. MF + Angle-Independent ASF Approximation

For Rayleigh scattering of an unpolarized photon, the differential cross section in modified relativistic form-factor plus anomalous-scattering-factor approximation is

$$\frac{d\sigma^{\text{MF+ASF}}}{d\Omega} = \frac{r_0^2}{2} |g(q) + g'(\omega, \theta) + ig''(\omega, \theta)|^2 (1 + \cos^2\theta) \ , \tag{1}$$

where θ is the scattering angle ($\theta = 2\Theta$, where Θ is the Bragg angle), and r_0 is the classical electron radius. The total-atom modified form factor is given as

$$g(q) = \sum_n g_n(q) = 4\pi \sum_n \int_0^\infty \rho_n(r) \frac{\sin qr}{qr} \left[\frac{mc^2}{E_n - V(r)} \right] r^2 dr \ , \tag{2}$$

where the sum n is over the bound electrons of the atom, and g', g'' are the anomalous scattering factors associated with the modified form factor, g.

For forward angles, the anomalous scattering factors defined above satisfy the dispersion relation

$$g'(\omega, 0) = \frac{2}{\pi} \int_0^\infty \frac{\omega' g''(\omega', 0)}{\omega'^2 - \omega^2} d\omega' \ , \tag{3}$$

and the optical theorem

$$g''(\omega, 0) = - \frac{\omega}{4\pi c r_0} \sigma^{\text{TOT}} \ , \tag{4}$$

where σ^{TOT} is the total photo-atom cross section, ω is the photon frequency, and c is the speed of light. Although the total cross section at x-ray energies is dominated by

absorption, primarily atomic photoeffect, contributions from other processes must be included if accurate results are to be obtained, as is discussed subsequently.

Our numerical calculations have shown that the finite-angle anomalous scattering factors defined above are insensitive to the scattering angle θ for photon energies below the K-shell threshold. And although our numerical calculations show that the anomalous scattering factors exhibit angle dependence, especially for high momentum transfer for high Z, we will utilize the approximation of angle-independent anomalous scattering factors for all energies as we have not yet discovered a simple expression for modeling the angle dependence of anomalous scattering factors that is a significant improvement for all Z, energies and angles. That is, we assume

$$g'(\omega, \theta) = g'(\omega) , \quad g''(\omega, \theta) = g''(\omega) . \tag{5}$$

We are currently exploring use of analytic Coulomb K-shell predictions [13] as a means of providing improved angular distributions.

The anomalous scattering factors g', g'' , which are zero in the limit of high energy, are closely related to the anomalous scattering factors f', f'' conventionally defined in reference to the nonrelativistic high-energy limit for the forward-angle elastic scattering amplitude, $-Nr_0$ (N is the number of bound electrons in the atom), as

$$g'(\omega) = f'(\omega) - f'(\infty) , \quad g''(\omega) = f''(\omega) . \tag{6}$$

Kissel and Pratt [4] provide an improved estimate for the high-energy limit and corrections

that should be applied to the ASF tabulations of Cromer and Liberman [8] and the older tabulations of Henke et al. [9]; the most recent ASF tabulation of Henke et al. [10] already include these corrections.

Starting with the same total photo-atom cross sections, the forward-angle elastic-scattering cross section computed using g', g'' will be the same as that evaluated using f', f''. But these two approaches are not equivalent at finite angles. The angle-independent g', g'' form is a better choice for high-energy, large-angle scattering (i.e., high momentum transfer) for high Z because the finite high-energy limit of f', although small compared with the forward-angle value of $f(0)$, dominates the real part of the scattering amplitude at large angles giving very poor predictions.

3. Partitioning the Elastic Scattering Amplitude

For elastic scattering, we have shown that, starting with the many-body scattering amplitude and partitioning the scattering amplitude into Rayleigh and Delbrück scattering components, contributions from processes beyond the familiar terms for photoionization need to be included [12]. To lowest order in the fine structure constant, the total cross section used in the optical theorem for Rayleigh scattering is

$$\sigma^{TOT} = \sigma^{PE} + \sigma^{BBT+} - \sigma^{BBT-} - \sigma^{BPA} , \qquad (7)$$

where σ^{PE} is the cross section for photoeffect, σ^{BBT+} is the cross section for a transition from the initial state to a excited bound state of the system, σ^{BBT-} is the cross section for a transition from the initial state to a bound state of lower energy, and σ^{BPA} is the cross

section for bound-electron pair annihilation wherein an initial hole in the negative-energy sea is filled by one of the initial bound electrons.

Only with the inclusion of all the contributions in Eq. (7) is: 1) the dispersion integral Eq. (3) convergent (both σ^{PE} and σ^{BPA} decrease as $1/E$ at high energies); 2) agreement achieved for forward-angle predictions of the anomalous-scattering-factor approximations with the more sophisticated S-matrix approach; 3) the Thomas-Reiche-Kuhn sum rule satisfied.

It has generally been thought of as small compared with the contribution from bound-free transitions, and therefore bound-bound transitions have been ignored in published anomalous scattering tables [8-10]. It was shown some time ago [14] that for atoms or ions with less than ten remaining bound electrons, the contribution from bound-bound transitions, especially from the *1s → 2p* transition, is significant and these transitions should be included in the real anomalous scattering factor, g' or f'. The inclusion of bound-bound transitions is important for satisfying the relativistic Thomas-Reiche-Kuhn (TRK) sum rule [15]. The effect of neglecting bound-bound transitions is especially significant for lower photon energies, not only for atoms with less than 10 bound electrons but also for other atoms in general. Bound-bound transitions contribute 30% or more to the sum rule for atoms $Z = 1$, 3-5, 20, 21 and 56-58.

4. New ASF Predictions

Using a Dirac-Slater IPA model for the atom, we have prepared new tabulations of anomalous scattering factors (ASF) for all Z=1-99, for photon energies 0-10,000 keV. These tabulations have been prepared with a variable energy grid to support 2-point linear

interpolation accurate to a fraction of a percent. In our evaluation of the ASF, we have made essentially no approximation in the evaluation of the photoeffect cross section at all energies 0-100,000 keV. We directly evaluate bound-bound oscillator strengths for the most significant bound-bound contributions to g' (over 500 transitions were included for C; over 1500 transitions were computed for Pb). In our evaluation of Eq. (3), we continued the integration above the pair-production threshold $2mc^2$ to an energy of 100 MeV. (Our anomalous scattering factors at about 1 MeV were insensitive to cutoffs above about 15 MeV.) The total-atom cross sections above $2mc^2$ were obtained using analytic semi-relativistic expressions [16].

These ASF calculations have been extended to zero energy to check how well our values satisfy the TRK sum rule (to a fraction of a percent in most cases) and to provide support for radiation transport calculations for all energies, even though the validity of our IPA atomic model becomes increasingly poor.

5. Future Efforts

Our Rayleigh-scattering calculations have been performed for an isolated, neutral, ground-state atom within the framework of IPA. This approach is insufficient when correlations among electrons become important (such as for very low photon energies or very close to thresholds) or when the atom is strongly affected by an environment (such as in the solid state or a plasma).

Further efforts in incorporation of correlation effects can proceed by using one of several existing approaches such as many-body perturbation theory [19] or random phase approximation [20]. The recent tabulation of ASF due to Henke et al. [10] includes some

electron correlations through use of time-dependent local-density approximation photoabsorption data [21] at low energies.

The anomalous-scattering-factor approach provides opportunities to incorporate experimental information, such as experimental atomic thresholds and near-threshold photoabsorption data, into practical tabulations. In an initial demonstration [3], we have utilized experimental near-edge photoabsorption data, coupled to Dirac-Slater IPA photoabsorption data away from the edge, to compute near-edge anomalous scattering factors. These mixed theoretical/experimental ASF values show a dramatically improved agreement with the near-edge ASF measurements.

Our ASF calculations utilize photoabsorption data computed from a Dirac-Slater IPA model, which exhibits differences in detail with experiment. We have used an energy-shift algorithm to incorporate experimental information in our ASF calculations that enforces the experimentally observed edges in photoabsorption data [17]. The resulting ASF values demonstrated a vast improvement over unshifted ASF values with recent near-edge experimental observations [18] based on mirror reflectivity measurements. The energy-shifted ASF values still satisfy the TKR sum rule to order of 1% or better.

The angle-independent assumption for ASF fails for finite angles at photon energies well above the K threshold. Recent investigations of multipole and retardation effects for scattering from the ground state of hydrogen [13] yield analytic expressions for the angle dependence of the anomalous scattering factor. Further efforts to incorporate these new results into our numerical ASF calculations are warranted.

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